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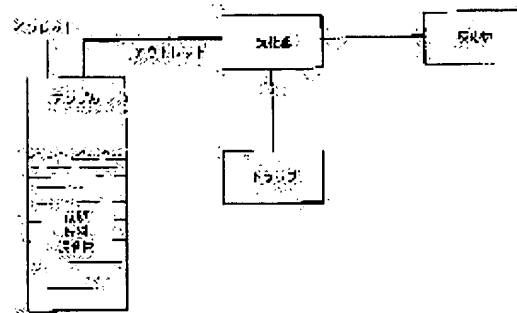
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(54) PRECURSORY RAW MATERIAL MIXTURE, FILM DEPOSITION METHOD AND FORMATION OF STRUCTURE

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a precursory raw material mixture useful for CVD and ALD, to provide a method for growing a film by using the same and to provide a method for forming an electronic device having the same film incorporated therein.

SOLUTION: At least one precursory compound containing a metallic element such as Li contains at least one precursory material inclusive of a compound such as a hydride, and the same is dissolved, emulsified or floated into an inert solution of aliphatic hydrocarbon or the like. The precursory raw material mixture is a solution, an emulsion or a suspension and is composed of a mixture of a solid phase, a liquid phase and a vapor phase, and they are dispersed over the whole mixture.



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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] Especially this invention relates to useful precursive raw material mixture about the adhesion method in a chemical vacuum deposition (CVD:chemical vapor deposition) and an atomic-layer adhesion (ALD:atomic layer deposition) process. this invention relates to the method of forming the electronic device which includes at least one film to which it adheres from the precursive raw material mixture of this invention, a layer, or covering again.

[0002]

[Description of the Prior Art] If the size of a semiconductor device contracts in order to improve a performance, the need of managing thickness in a thinner uniform size will increase. When a size contracts, the homogeneity of thickness is important in order to minimize a roll with a contiguity element. The homogeneous improvement is important in the present technology of using the \*\* source / drain diffusion (0.25 micrometers). Since shallow diffusion is gained by placing which leads an oxide, the heterogeneity of the thickness of the oxide produces uneven source / drain diffusion depth, and it degrades an element performance. Between etching, the homogeneous improvement of thickness is important again in order to minimize the over etching which is directly dependent on membranous homogeneity.

[0003] in semiconductor application, in order for almost all the films to which CVD or ALD adheres to grow using the conventional bubbler (bubbler) technology of being an elevated temperature and whipping carrier gas through a raw precursor (that is, there being no solvent) and to surround a film with uniform precursive flux in this case, it is desired for the vapor pressure of a precursor to be fixed however, since vapor pressure is directly related to temperature, the conventional bubbler technology has the fault that it is required that the Barbra temperature should be maintained to the minimum change between runs or between a run and a run It is known that change of precursive flux will lead to change of a film growth rate. It sinters in connection with the passage of time, changing a surface field is known, and an individual compound produces the ununiformity of the film growth rate between runs. Although sintering is not a problem for a liquid precursor, along with the passage of time, a liquid precursor may deteriorate with the heat cycle and thermal load which are imposed on it. Furthermore, a decomposition process is accelerated at an elevated temperature. The elevated temperature of the precursor between evaporation within the conventional bubbler and a heat cycle contribute to early degradation of a precursor accompanying the passage of time. A precursor changes those chemistry states by the ligand rearrangement, cluster formation, or oxidization. A precursor can react with the water or oxygen which may be carelessly introduced in a bubbler through the water and oxygen which are absorbed on the carrier gas inadequate in purification foam-ized through a precursor, and an air leak or a bubbler wall.

[0004] generally in the conventional Barbra technology, it is used, and a hydride, an alkyl, ARUKENIRU, cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, a phosphide, a nitrate, a halogenide, an alkoxide, siloxide, and silyl are contained in the example of the precursor which wears above-mentioned un-arranging Especially the unstable hydride is thermally [ alkylamine Alan etc. ] attractive for CVD and ALD. Those high reactivity is because impurity mixing is usually reduced through low heat treatment temperature. Though regrettable, alkylamine Alan is unstable as everyone knows between preservation, a transfer, and evaporation, consequently membranous repeatability is bad. Decomposing alkylamine Alan, such as trimethylamine Alan, triethylamine Alan, and diethyl monomethylamine Alan, above 40 degrees C between preservation and between the transfers to a CVD reactor is found. Cautions are required, in order to save a precursor below at a room temperature and to minimize decomposition. Therefore, a transfer and evaporation temperature are restricted by the pyrolysis of a precursor (1994 for example, Dario M.Frigo and "Chemistry of Materials" by Gerbrand J.M.van Eijden, 6,190 -195, C.E.Chrysosou and "Applied Physics A Materials Science and Processing" by C.W.Pitt, vol. 65, 1997, 469 -475 reference).

[0005] There are Cu (I) compounds, such as Cu (cyclopentadienyl) (PET3), in another example of an unstable precursor thermally, this is about 70 degrees C in low temperature, and losing PET3 and decomposing is known. There are alkyls, such as trimethylindium and triethylindium, in other examples. Triethylindium is a liquid and decomposing at a room temperature in a bubbler is found. At a room temperature, trimethylindium is a solid-state, and in connection with the passage of time, change of effective vapor pressure is observed and it produces the heterogeneity which is not desirable and the non-repeatability of a growth result (G. refer to "Organometallic Vapor-Phase Epitaxy:Theory and Practice" by B.Stringfellow, San Diego, CA:Academic Press, and 1989).

[0006] An alkoxide is contained in other examples and it is known that this will change a chemistry state by the ligand rearrangement accompanying the passage of time, hydrolysis, co-oligomerization, ring formation, cluster formation, and oxidization. These decomposition processes are accelerated in the elevated temperature which encounters with the conventional bubbler

technology. Furthermore, especially the alkoxide is sensitive to water and an oxygen impurity, and these may be carelessly introduced in a bubbler through the water and oxygen which are absorbed on the carrier gas inadequate in purification air-bubbled through a precursor, and an air leak or a bubbler wall. A adding-water decomposition reaction may occur and these reactions are accelerated in the elevated temperature which generally encounters in the conventional bubbler technology. An alkoxide may exist with many isomer forms which carry out mutual change with time again, consequently produces adjustable vapor pressure. For example, aluminum isopropoxide exists with many isomer forms with the mutual change speed slowly carried out between isomers. It is made difficult that it controls the sticking rate of aluminum  $2O_3$  which grows from this compound using the conventional bubbler technology since the vapor pressure of these isomers changes a lot (R. G.Gordon, K Kramer, X. MRS Symp Proc.Vol. by Liu 446, 1997, and p.383 reference).

[0007] There is an amide served like an alkoxide in other examples, and this has a ligand rearrangement, hydrolysis, oxidization, co-oligomerization, and the inclination of ring formation, exists with the isomer form in which some mutual change is possible, and generates unreproducible vapor pressure in connection with the passage of time. Non-liquid gold group nitrates, such as nitric-acid titanium, a zirconium nitrate, and a nitric-acid gallium, are contained in other examples. These complexes are sensitive to air and water, and decomposing at the temperature of about 100 degrees C is known. In addition to being sensitive to air and water, metal oxo nitrates, such as  $VO(NO_3)_3$  and  $CrO_2(NO_3)_2$ , are sensitive to light, and should be saved at 0 degree C. "Chemical Vapor Deposition" according to D.G.Colombo, D.C.Gilmer, V.G.Young, S.A.Campbell, and W.L.Gladfelter for example about this, 1998 and 4, and No. -- it is indicated by 6, 1998, and P.220

[0008]

[Problem(s) to be Solved by the Invention] It is already said that beta diketonate containing the precursor dissolved into the solution for CVD growth is used. U.S. Pat. No. 5204314, 5225561, 5280012, 5453494, and 5919522 are indicating about the method of growing up calcium, Sr, or Ba, using the solution containing calcium, Sr, or Ba complex combined with at least one beta diketonate ligand or beta diketonate derivative. U.S. Pat. No. 5555154 is indicating about growth of  $PbZrTiO_3$  by the chemical vacuum deposition in the case of using the solution containing Pb, Zr, and Ti dipivaloyl meta-NETO in a tetrahydrofuran. U.S. Pat. No. 5677002 and 5679815 indicate about the method of growing up the tantalum and niobium containing a film, using the solution of Nb and Ta which are combined with at least one beta diketonate ligand or beta diketonate derivative. U.S. Pat. No. 5698022 is teaching useful precursor composition to the chemical vacuum deposition of a lanthanide metal / phosphoric-acid ghost film, and this contains the precursor compound which consists of the lanthanide metal beta diketonate and the Lynn content ligand in a solvent. U.S. Pat. No. 5783716 indicates about the method of growing up Pt by CVD, using the solution containing Pt complex combined with at least one beta diketonate ligand or beta diketonate derivative. U.S. Pat. No. 5820664 is indicating about the useful metal raw material reagent solution to the chemical vacuum deposition, and this contains the metal coordinated complex containing the metal by which coordinate bond is carried out to at least one beta diketonate ligand or beta diketonate derivative. U.S. Pat. No. 5900279 indicates the solution which consists of beta diketonate content precursor dissolved into one of the ligands of a complex. U.S. Pat. No. 5916359 indicates the method of growing up  $SrBi_2Ta_2O_9$  by CVD, using the precursor composition containing beta diketonate content precursor of Sr, Bi, and Ta dissolved into two C6-C12 different alkanes, the solvents of the Grimm base, or 3 component solutions of polyamine. U.S. Pat. No. 5980983 indicates use of the mixture of metal beta diketonate for adhesion of a metal content film. In spite of various indications of beta diketonate precursor, it is known that beta diketonate content precursor has a complex decomposition path, and, thereby, the carbon of a considerable amount or other unnecessary impurities are built into the film of a result.

[0009] U.S. Pat. No. 5900279 is teaching the solution useful for CVD, and this contains the metal organic compound added to the liquid which consists of the ligand of a metal organic compound fundamentally. For example, M (beta diketonate) is dissolved within beta diketone. This quotation wears the fault to which a superfluous ligand exists between decomposition of the precursor for film formation. A ligand solvent has the inclination to take the same decomposition path as a precursor and precursor decomposition fragmentation, therefore is in a gaseous phase or bars decomposition of the precursor on the front face of a film. The gaseous phase reaction between the evaporated precursor, the precursor decomposed partially, the evaporated ligand solvent, and its decomposition by-product may occur, and particle formation within volatile deterioration of a precursor, a carburetor, and a reactor and an unreproducible growth rate as a result may be produced.

[0010] In order to form the layer or film which there is the need of developing an adhesion process continuously, in view of the fault about the conventional adhesion process, and is used in various electronic devices and to which it adhered thinly, it is desirable to use the newly improved precursive raw material mixture.

[0011] this invention relates to the method of growing up a film (they being a layer, covering, and a multilayer to this appearance) using useful precursive raw material mixture and the precursive raw material mixture of this invention for CVD and ALD application, and the method of forming the electronic device incorporating the film to which this invention adhered. A transistor, a capacitor, diode, a resistor, a switch, light emitting diode, laser, wiring structure, interconnection structure, or other arbitrary structures where the film of this invention is incorporated are included in the suitable electronic device formed of this invention.

[0012]

[Means for Solving the Problem] Especially the precursive raw material mixture of this invention Li, Na, K, Rb, Cs, Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, aluminum, At least one precursor which consists of the element chosen from the group containing Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi is included. To this, a hydride, an alkyl, ARUKENIRU, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, At least one ligand chosen from the group containing nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative is combined. A ligand dissolves, emulsifies or floats in aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a

halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, an alkyl nitrate, and the inactive liquid chosen from the group containing such mixture. Precursive raw material mixture is a solution, an emulsion (emulsion), or a suspension (suspension), it consists of mixture of solid phase, the liquid phase, and a gaseous phase, and these are distributed over the whole mixture.

[0013] this invention relates to CVD or the ALD method for growing up a film on a substrate again using the precursive raw material mixture of this invention. The film formation method contains the step which evaporates the precursor in precursive raw material mixture, and the step which adheres the component of the evaporated precursor on a substrate and forms a film. It does not need to be evaporated even if inactive liquid is evaporated together with a precursor about this point. In the one example, inactive liquid is evaporated together with a precursor. In the another example of this invention, inactive liquid is not evaporated but is transmitted with the form of a liquid from a reactor.

[0014] Another mode of this invention is related with formation of the multilayer structure incorporating one or more layers to which it adheres by this method.

[0015] Furthermore, another mode of this invention is related with formation of the compound element film (multicomponent film) with which at least one component is drawn from the precursive raw material mixture of this invention.

[0016] Furthermore, the following are contained in another mode of this invention. Namely, formation of the electronic structure incorporating the film to which it adheres by this method. Formation of the complementary-metal-oxide-semiconductor (CMOS) integrated-circuit logical element which incorporates the film to which it adheres by this method, and contains both n type field-effect transistor (NFET) formed on a single substrate, and p type field-effect transistor (PFET) as shown in drawing 1. Formation of the integrated circuit capacitor which incorporates the film to which it adheres by this method as shown in drawing 2. Formation of the integrated-circuit wiring structure which incorporates the film to which it adheres by this method as shown in drawing 4.

[0017]

[Embodiments of the Invention] As mentioned above, this mixture contains at least one precursor and ii inactive liquid of i this invention about precursive raw material mixture with this invention useful to CVD or ALD.

[0018] A precursor Li, Na, K, Rb, Cs, Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, aluminum, Ga, In, Tl, Si, It defines as arbitrary compounds containing the element chosen from the group containing germanium, Sn, Pb, As, P, Sb, and Bi. To this, a hydride (H), an alkyl (CR<sub>3</sub>), ARUKENIRU (CRCR<sub>2</sub>), Cyclo ARUKENIRU, an aryl, an alkyne (CCR), a carbonyl (CO), An amide (NR<sub>2</sub>), imide (NR), a hydrazide (NRNR<sub>2</sub>), a phosphide (PR<sub>2</sub>), A nitrosyl (NO), nitril (NO<sub>2</sub>), a nitrate (NO<sub>3</sub>), nitril (RCN), An isonitrile (RNC), a halogenide (F, Cl, Br, or I), At least one ligand chosen from the group containing an azide (N<sub>3</sub>), alkoxy (OR), siloxy (OSiR<sub>3</sub>), silyl (SiR<sub>3</sub>) and these halogenation, sulfonation, or a silicification derivative is combined. And if sent to a carburetor, a precursor will be easily changed into a gas. The halogenation derivative of a ligand is defined as substitution of H substituent by the halogen chosen from the group containing F, Cl, Br, and I. The sulfonation derivative of a ligand is defined as substitution of O substituent by S. The silicification derivative of a ligand is defined as substitution of C substituent by Si.

[0019] The generalization chemical formula of the precursor of this invention is expressed as follows.

[Formula 3] MR<sub>1</sub>xR<sub>2</sub>yAz[0020] M here Li, Na, K, Rb, Cs, Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, It is the element chosen from the group containing Ag, Au, Zn, Cd, Hg, B, aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi. R<sub>1</sub> and R<sub>2</sub> A hydride, an alkyl, ARUKENIRU, cyclo ARUKENIRU, an aryl, an alkyne, A carbonyl, an amide, imide, a hydrazide, a phosphide, a nitrosyl, Nitril (nitril), a nitrate, nitril (nitrile), it is chosen from the group containing an isonitrile, a halogenide, an azide, alkoxy \*\*, siloxy, silyl and these halogenation, sulfonation, or a silicification derivative -- said -- 1 -- or it is a different ligand A is arbitrary ligands which join together or meet in coordination. Moreover, a phosphine (R<sub>3</sub>P), Phosphite ((RO)<sub>3</sub>P), an amine (R<sub>3</sub>N), an arsine (R<sub>3</sub>As), A stibine (R<sub>3</sub>Sb), the ether (R<sub>2</sub>O), a sulfide (R<sub>2</sub>S), nitril (RCN), It is chosen from an isonitrile (RNC), an alkene, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol (ROH), a phosphinic acid ghost (R<sub>3</sub>PO), alkylidene, a nitrite, an alkyne, and the group containing water. Moreover, it is the valence of x>=1 and the x+y= element M, and is y>=0 and z>=0.

[0021] The suitable precursor of this invention is a compound evaporated easily. Especially a suitable precursor is dimethyl -, diethyl -, or diisobutyl - B, aluminum, Ga, In, As, or Sb hydride; Me<sub>2</sub>AlH(NEtMe<sub>2</sub>); -- the 3rd -- butyl arsine; (Me<sub>3</sub>N) AlH<sub>3</sub>; (EtMe<sub>2</sub>N) AlH<sub>3</sub>; (Et<sub>3</sub>N) AlH<sub>3</sub>; CpWH<sub>2</sub>; Cp<sub>2</sub>MoH<sub>2</sub>; TORIMECHIRU -, triethyl -, and TORIISO butyl - TORI n propyl -, triisopropyl -, TORI n butyl -, TORINEO pentyl -, Or ethyl dimethyl-B, aluminum, Ga, In, As or Sb; Tetramethyl -, Tetraethyl -, tetrapod phenyl -, or tetrapod n butyl-Si, germanium, Sn, or Pb; dimethyl -, diethyl -, or diisobutyl - B, aluminum, Ga, In, As or Sb hydride, a chloride, a fluoride, a bromide, An iodide, Cp, an amide, a dimethyl amide, or an azide; Triethyl -, TORIISO butyl -, TORI n propyl -, triisopropyl -, TORI n butyl -, Or ethyl dimethyl - B, aluminum, Ga, In, As, or Sb trimethylamine, A diethyl monomethylamine, a dimethyl ethylamine, or a triethylamine; Dimethyl - or diethyl-Zn, Cd or Hg; (neopentyl) 4 Cr; Et<sub>3</sub>Pb(neo pentoxy); Cp<sub>2</sub>Me<sub>2</sub>Zr; (MeNC) 2PtMe<sub>2</sub>; CpIr<sub>2</sub> (C<sub>2</sub>H<sub>4</sub>); Screw Cp-Co, Mo, Fe, Mn, nickel, Ru, V, Os, Mg, or Cr; screw ethylbenzene; screw benzene - Co, Mo or Cr; triphenyl - Bi, Sb, or As; TORIBI nil boron; tris Cp-Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb Or Lu; tris allyl-compound iridium; CpCr (CO) 2; Cp<sub>2</sub>ZrMe<sub>2</sub>; CpCuPt<sub>3</sub>; EtCpCuPt<sub>3</sub>; CpIn; CpIr; CpPd (Cyclo-octadiene); CpGaMe<sub>2</sub>; CpGaEt<sub>2</sub>; (Allyl compound) FeCO<sub>3</sub>; (Cyclohexadiene) FeCO<sub>3</sub>; ethyl ferrocene; CpMn(CO)<sub>3</sub>; (cycloheptatriene) Mo(CO)<sub>3</sub>; NdCp<sub>3</sub>; SmCp<sub>3</sub>; ScCp<sub>3</sub>; TbCp<sub>3</sub>; TiCp; TmCp<sub>3</sub>; Cp<sub>2</sub>WH<sub>2</sub>; (mesitylene) W (Cyclo-octatriene) (CO) 3; CpRe (CO) 3; CpRh (CO) 2; Ir (Allyl compound) 3; P t 2; CpIr (Allyl compound); [Ir (Cyclo OKUTA screw-on) (OMe) ] 2; Ru (Cyclo OKUTA screw-on) (Cyclo OKUTA screw-on) 2; Ru<sub>3</sub>CO<sub>12</sub>; Fe (Allyl compound) (CO) 5; Co<sub>2</sub> (CO) 8; Ru (CO) 3; Os<sub>3</sub>CO<sub>12</sub>; Cr (1, 3-cyclohexadiene) (CO) 6; CpCo (CO) 2; Mn<sub>2</sub> (CO) 10; CpMn (CO) 3; Mo (CO) 6; nickel (CO) 4; Re<sub>2</sub> (CO) 10; CpRe (CO) 3; CpRh (CO) 2; Ru<sub>3</sub> (CO) 12; W (CO) -- 6; CpV(CO)<sub>4</sub>; CF<sub>3</sub>Co [ ; P ] (CO) 4 t(CO) 2 (cyclo OKUTA screw-on); Ir(CO) 2 (cyclo OKUTA screw-on);

(CO) -- four -- Fe[P(OCH<sub>3</sub>)<sub>3</sub>];(CO)<sub>4</sub>Fe[N(CH<sub>3</sub>)<sub>3</sub>];CoNO(CO)<sub>3</sub>; -- butoxy one -- OCH(CF<sub>3</sub>)<sub>2</sub>, OCMe<sub>2</sub>(CF<sub>3</sub>), OCMe(CF<sub>3</sub>)<sub>2</sub>, OSi(CH<sub>3</sub>)<sub>3</sub>, OC(CH<sub>3</sub>)<sub>3</sub>, OC(SiMe<sub>3</sub>)<sub>3</sub>, or OC(CF<sub>3</sub>)<sub>3</sub>Li, Na, K, Rb, Cs, Fr, Cu, Ag, Au, Hg, or Tl; Tetramethoxy, Tetrapod ethoxy \*\* tetrapod isopropoxy, tetrapod butoxy, tetrapod n butoxy, Tetrapod iso butoxy, 3rd butoxy ones of a tetrapod, a tetrapod OCH(CF<sub>3</sub>)<sub>2</sub>, a tetrapod OCMe<sub>2</sub>(CF<sub>3</sub>), A tetrapod OCMe(CF<sub>3</sub>)<sub>2</sub>, a tetrapod (CH<sub>3</sub>) OC<sub>3</sub>, a tetrapod (SiMe<sub>3</sub>) OC<sub>3</sub>, a tetrapod (CF<sub>3</sub>) OC<sub>3</sub>, or tetrapod OSi(CH<sub>3</sub>)<sub>3</sub>Si, germanium, Sn, Pb, Ti, Zr or Hf;VO(isopropoxy)<sub>3</sub>, triisopropoxy, 2nd butoxy one of TORI, TORI n butoxy, TORIISO butoxy, trimethoxy, TORIETOKISHI, 3(OCH<sub>2</sub>(CF<sub>3</sub>)), 3(OCMe<sub>2</sub>(CF<sub>3</sub>)), 3(OCMe<sub>2</sub>(CF<sub>3</sub>)), 3(OC<sub>3</sub>(CH<sub>3</sub>)), 3(OC<sub>3</sub>(SiMe<sub>3</sub>)), 3(OC<sub>3</sub>(CF<sub>3</sub>)) or (OSi<sub>3</sub>(CH<sub>3</sub>))<sub>3</sub>, B, aluminum, Ga, In, P, As or Sb;Et<sub>3</sub>Pb(isopropoxide);(3rd butoxy) CuPMe<sub>3</sub>; Tetrakis(dimethylamino), Tetrakis Ti, Zr, Hf, Si, and germanium(dimethylamino), Sn or Pb; diethylamino diethyl arsine; -- diethylamino arsine dichloride; -- bisdimethyl amino arsine chloride; -- Me<sub>2</sub>Zn(triethylamine)<sub>2</sub>; diethylamino dimethyl stannane; -- tris(dimethylamino) phosphine; -- tris Antimony; Tris(Dimethylamino) It Li(s). (Dimethylamino) arsine; -- tris(dimethylamino) stibine; -- tris screw(trimethylsilyl) erbium amide; -- screw(dimethylamino)(trimethyl ethyl ethylene diamino) aluminum; -- (CO)<sub>4</sub>Fe[N(CH<sub>3</sub>)<sub>3</sub>]; -- Na Or KN CpCu(triphenyl phosphine); (SiMe<sub>3</sub>); -- PENTA dimethylamino tantalum; -- diethylamino dimethyl tin; -- hexa dimethylamino JITANGU stainless steel; -- tris dimethylamino(trimethylethylene diamino) titanium; -- (3rd butoxy) CuPMe<sub>3</sub>; P t 4;nickel(platelet factor 3) 4;Cr(platelet factor 3) 6;(platelet factor 3) 3Mo(Et<sub>3</sub>P)(CO)<sub>3</sub>;Ir 4;Ti(platelet factor 3) 4;Zr(NO<sub>3</sub>)<sub>4</sub>;Hf(NO<sub>3</sub>)<sub>4</sub>;Si(NO<sub>3</sub>)<sub>3</sub>(CH<sub>3</sub>); RuNO(NO<sub>3</sub>)(NO<sub>3</sub>)<sub>3</sub>; nitric-acid gallium; -- Sn(NO<sub>3</sub>)<sub>4</sub>;Co(NO<sub>3</sub>)<sub>3</sub>;VO(NO<sub>3</sub>)<sub>3</sub>;CrO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>;TiCl<sub>4</sub>;HfCl<sub>4</sub>;ZrCl<sub>4</sub>;InCl<sub>2</sub>;ZnCl<sub>2</sub>;AlCl<sub>3</sub>;SiCl<sub>4</sub> --;GaCl<sub>3</sub>;SnCl<sub>4</sub>;CoCl<sub>3</sub>; dimethyl - diethyl - or diisobutyl-aluminum, B, germanium and Si, or As halogenide;N(SiMe<sub>3</sub>)<sub>2</sub> -- Li, Na or K;B(CH<sub>2</sub>SiMe<sub>3</sub>)<sub>3</sub>;{(Me<sub>3</sub>Si)<sub>2</sub>-N}<sub>3</sub>-B, aluminum and Ga, or In; (Me<sub>3</sub>SiCH<sub>2</sub>)<sub>4</sub> - They are Ti, Zr or Hf;{(Me<sub>3</sub>Si)<sub>2</sub>-N}<sub>2</sub>-Zn, and Cd or Hg. Cp is cyclopentadienyl or alternative cyclo pen TAJINIERU, and the substitution of H substituent by a methyl, ethyl, an isopropyl, n butyl, the 2nd butyl, the 3rd butyl, trimethylsilyl, or other analogous substituents is taken into consideration here.

[0022] Between the precursive conservation of matter of this invention, and evaporation, inactive liquid is defined as arbitrary liquids which are not disassembled, when contacting a precursor. The inactive liquid used within precursive raw material mixture is chosen more as a detail from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C<sub>5</sub>-C<sub>12</sub> alkanes fundamentally. It is defined by capacity as 70% or 100% by this invention "is constituted fundamentally" here. Although an arbitrary additive may exist, an additive occupies about 30% of inactive liquid by capacity.

[0023] Selection of inactive liquid is based on the following criteria. That is, it is having inactive [ which is not decomposed between evaporation of a precursor / sufficient ], when contacting a precursor, and an unnecessary impurity is not introduced in the film of a result, without adsorbing disadvantageously for the film front face which grows, when this passes the pyrosphere of a reactor.

[0024] The precursive raw material mixture of this invention is a solution, an emulsion (emulsion), or a suspension (suspension), it consists of mixture of solid phase, the liquid phase, and a gaseous phase, and these are distributed over the whole mixture.

[0025] The precursive raw material mixture of this invention may be used with the arbitrary delivery meanses used now in arbitrary CVD(s) or ALD processes. Therefore, this invention is not limited to specific CVD or specific ALD equipment, or arbitrary delivery systems. A chemical vacuum deposition (CVD) is defined as a cotransduction of two or more reagents to a reactor. On the other hand, atomic-layer adhesion (ALD) is defined as sequential introduction of two or more reagents to a reactor, for example, atomic layer epitaxy, digitization study vacuum evaporation, a pulse chemical vacuum deposition, and other analogous methods are included.

[0026] According to this invention, in arbitrary CVD(s) or ALD processes, a film is formed on a substrate using the precursive raw material mixture of this invention. A film evaporates the precursor in precursive raw material mixture, and is formed by adhering the component of the evaporated precursor on a substrate after that. It does not need to be evaporated even if inactive liquid is evaporated together with a precursor about this point. In the one example of this invention, inactive liquid is evaporated together with a precursor. In the another example, inactive liquid is not evaporated but is transmitted with the form of a liquid from a reactor.

[0027] In addition to membranous formation, precursive raw material mixture is used in formation of the compound element film (multicomponent film) with which at least one component is drawn from the precursive raw material mixture of this method in formation of the multilayer structure incorporating one or more membrane layers of this invention.

[0028] Moreover, the precursive raw material mixture of this invention is used in formation of the structure incorporating the film to which it adhered by this method, i.e., electronic device structure. By this invention, since a transistor, a capacitor, diode, a resistor, a varistor, a switch, light emitting diode, laser, wiring structure, and interconnection structure are meant, term" electronic device structure" is used.

[0029] Furthermore, precursive raw material mixture is used in formation of a complementary-metal-oxide-semiconductor (CMOS) integrated-circuit logical element. this invention relates to formation of the CMOS integrated circuit containing both n type field-effect transistor (NFET) formed on a single substrate, and p type field-effect transistor (PFET) more at a detail. As shown in drawing 1, the gate electrode 14 by which the NFET element 11 is formed on p type conductivity field 13 of a substrate 10, and is formed on the gate dielectric 15, and one pair of n type sources / drain fields 16 which are formed in the both sides of the longitudinal direction of the gate electrode 14 are included. The gate electrode 19 by which similarly the PFET element 17 is formed on n type conductivity field 18 of a substrate 10, and is formed on the gate dielectric 15, and one pair of p type sources / drain fields 20 which are formed along with the side attachment wall of the both sides of the gate electrode 19 are included. NFET and a PFET element are separated by the \*\* trench separation 21 and the spacer 22. It adheres to at least one of the transistor components containing the gate electrode 14, the gate electrode 19, the gate dielectric 15, and a spacer 22 by this method about this point of this invention.

[0030] The precursive raw material mixture of this invention is used for formation of an integrated circuit capacitor again. As shown

in drawing 2, a typical capacitor is formed together with a barrier 32 on a substrate 30, and is connected to a transistor by the plug 31. A capacitor contains the bottom electrode 33, a dielectric (a ferroelectricity or non-ferroelectricity) 34, and the top electrode 35. It adheres to at least one of the capacitor components containing a plug 31, a barrier 32, the bottom electrode 33, a dielectric 34, and the top electrode 35 by this method about this point of this invention. A capacitor may be a stack or a trench.

[0031] Precursive raw material mixture is used in formation of integrated-circuit wiring structure again. As shown in drawing 3, typical wiring structure is formed by \*\*\*\*\*ing a trench 41 and Bahia 42 into a dielectric layer 43. Under a dielectric layer 43, the metal thin film wiring 44 and the dielectric layer 45 of a wiring layer exist. A trench and Bahia are filled up with drawing 4 with the barrier material 46 and the wiring metal 47. It adheres to at least one of the wiring structure components containing dielectric layers 43 and 45, the metal thin film wiring 44, the barrier material 46, and the wiring metal 47 by this method about this point of this invention.

[0032] The barrier layer which \*\*-form-covers the etching feature of double pellet scene structure is also formed using the precursive raw material mixture of this invention.

[0033] The aforementioned explanation is all-inclusive explanation of this invention, and, below, specific detail of this invention will be given.

[0034] Precursive raw-material mixture for a hydride content compound: The suitable precursive raw material mixture of a hydride content compound is constituted as follows.

It Fr(s). i) --  $MR_1xR_2yAz$  -- here -- M -- Li, Na, K, Rb, and Cs -- Be, Mg, Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Cu, It is the element chosen from the group containing Ag, Au, Zn, Cd, Hg, B, aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, Sb, and Bi, and they are B, aluminum, Ga, In, As, Sb, Si, germanium, Sn, Pb, Zn, Cd, and Hg suitably. Moreover, R1 may be a hydride, and R2 may be a ligand chosen from the group containing a hydride, an alkyl, ARUKENIRU, cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, a hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same A is arbitrary ligands by which coordinate bond is carried out, and is chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0035] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0036] In order to improve the stability of a compound in the case of an uneasy hydride content compound with the inclination decomposed between iii arbitrary additive preservation or evaporation, an additional non-hydride ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of a hydride content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0037] In the especially suitable precursive raw material mixture which consists of at least one hydride content compound, it is dimethyl -, diethyl -, or diisobutyl. - B, aluminum, Ga, In, As, Sb hydride; 3rd butyl arsine; CpWH<sub>2</sub>, or Cp<sub>2</sub>MoH<sub>2</sub> is contained, and these dissolve, emulsify or float fundamentally in the liquid which consists of C5-C12 alkane liquid. Other suitable precursive raw material mixture consists of at least one hydride content compound. This contains Me<sub>2</sub>AlH(NEtMe<sub>2</sub>);(Me<sub>3</sub>N) AlH<sub>3</sub>;(EtMe<sub>2</sub>N) AlH<sub>3</sub>, AlH<sub>3</sub>, etc. These emulsify or float in the liquid to which the amine was added alternatively (30% [ However, this capacity ] or less of inactive liquid) and which consists of C5-C12 alkane liquid fundamentally.

[0038] Precursive raw-material mixture for an alkyl content compound: The suitable precursive raw material mixture of an alkyl content compound is constituted as follows.

It Fr(s). i) --  $MR_1xR_2yAz$  -- here -- M -- Li, Na, K, Rb, and Cs -- Be, Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Cu, Ag, It is the element chosen from the group containing Au, Zn, Cd, Hg, B, aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, Sb, and Bi, and they are B, aluminum, Ga, In, As, Sb, Si, germanium, Sn, Pb, Zn, Cd, and Hg suitably. Moreover, R1 is C1-C8 alkyl or C4-C12 cycloalkyl. R2 A hydride, an alkyl, ARUKENIRU, cyclo ARUKENIRU, an aryl, An alkyne, a carbonyl, an amide, imide, a hydrazide, a phosphide, a nitrosyl, Nitril (nitril), a nitrate, nitril (nitrile), It may be the ligand chosen from the group containing a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0039] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0040] In order to improve the stability of a compound in the case of an uneasy alkyl content compound with the inclination



decomposed between iii arbitrary additive preservation or evaporation, an additional alkyl ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of an alkyl content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity. [0041] Into the especially suitable precursive raw material mixture which consists of at least one alkyl content compound TORIMECHIRU -, triethyl - or TORIISO butyl -, TORI n propyl -, Triisopropyl -, TORI n butyl -, TORINEO pentyl -, or ethyl dimethyl - B, aluminum, Ga, In, As, or Sb; Tetramethyl -, tetraethyl -, Tetrapod phenyl - or tetrapod n butyl-Si, germanium, Sn or Pb; Dimethyl -, Diethyl - or diisobutyl - B, aluminum, Ga, In, As, or Sb hydride, A chloride, a fluoride, a bromide, an iodide, Cp, an amide, a dimethyl amide, or an azide; Triethyl -, TORIISO butyl -, TORI n propyl -, triisopropyl -, TORI n butyl -, Or ethyl dimethyl - B, aluminum, Ga, In, As, or Sb trimethylamine, A diethyl monomethylamine, a dimethyl ethylamine, or a triethylamine; A dimethyl or Diethyl Zn Cd, Hg;(neopentyl) 4Cr;Et3Pb(neo pentoxy);Cp2Me2Zr;(MeNC) 2PtMe2;, or CpIr (C2H4)2 is contained. Cp is cyclopentadienyl or alternative cyclo pen TAJINIERU, and the substitution of H substituent by the methyl which dissolves, emulsifies or floats in C5-C12 alkane liquid, ethyl, an isopropyl, n butyl, the 2nd butyl, the 3rd butyl, trimethylsilyl, or other analogous substituents is taken into consideration here. Other suitable precursive raw material mixture consists of at least one alkyl content compound, and trimethyl \*\*\*\* which emulsifies or floats in the liquid to which this had methane or ethane added alternatively (30% [ However, these capacity ] or less of inactive liquid), and which consists of C5-C12 alkane liquid fundamentally contains triethyl etc.

[0042] Precursive raw-material mixture for an ARUKENIRU content compound: The suitable precursive raw material mixture of an ARUKENIRU content compound is constituted as follows.

It Fr(s). i) -- MR1xR2yAz -- here -- M -- Li, Na, K, Rb, and Cs -- Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, aluminum, It is the element chosen from the group containing Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are Bi, As, Cr, Zr, Cu, Co, In, Ir, Fe, La, Mg, Mn, Mo, nickel, Os, Ru, Tl, or W suitably. R1 Moreover, C1-C8 ARUKENIRU, C4-C12 cyclo ARUKENIRU, It is C5-C12 aryl. R2 Or a hydride, an alkyl, ARUKENIRU, Cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, It may be the ligand chosen from the group containing nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0043] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0044] In order to improve the stability of a compound in the case of an uneasy ARUKENIRU content compound with the inclination decomposed between iii arbitrary additive preservation or evaporation, an additional ARUKENIRU ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of an ARUKENIRU content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0045] Especially the suitable precursive raw material mixture that consists of at least one ARUKENIRU content compound Screw Cp-Co, Mo, Fe, Mn, nickel, Ru, V, Os, Mg or Cr; Screw ethylbenzene -, Screw benzene-Co, Mo or Cr; triphenyl - Bi, Sb, or As; TORIBI nil boron; tris Cp-Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, or Lu; Tetrapod CpTh, Pa, V, Np, Pu Or Am; tris allyl-compound iridium; CpCr (CO) -- 2;Cp2ZrMe2;CpCuPEt3;CpIn;CpIr(cyclo-octadiene);CpPd(allyl compound);CpGaMe2 -- ;CpGaEt2;(cyclohexadiene) FeCO3;(cyclo-octatriene) FeCO3; ethyl ferrocene; -- CpMn (CO) 3; Mo (Cycloheptatriene) (Co) 3;NdCp3;SmCp3;ScCp3;TbCp3;TlCp;Cp2WH2;(mesitylene) W(CO)3;CpRe(CO)3;CpRh(CO)2;Ir3 (allyl compound) ;P t(allyl compound)2;CpIr(cyclo OKUTA screw-on);[Ir It is chosen from the group containing]2; and Ru (cyclo OKUTA screw-on) (allyl compound)2. (OMe) (cyclo OKUTA screw-on) Cp is cyclopentadienyl or alternative cyclo pen TAJINIERU here. The substitution of H substituent by the methyl which dissolves, emulsifies or floats in C5-C12 alkane liquid, ethyl, an isopropyl, n butyl, the 2nd butyl, the 3rd butyl, trimethylsilyl, or other analogous substituents is taken into consideration.

[0046] Precursive raw-material mixture for a carbonyl content compound: The suitable precursive raw material mixture of a carbonyl content compound is constituted as follows.

It Fr(s). i) -- MR1xR2yAz -- here -- M -- Li, Na, K, Rb, and Cs -- Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, aluminum, It is the element chosen from the group containing Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are Ru, Fe, Co, Os, Cr, Mn, Mo, nickel, Re, Rh, W, Pt, or Ir suitably. R1 is a carbonyl. R2 Moreover, a hydride, an alkyl, ARUKENIRU, Cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, It may be the ligand chosen from the group containing nitril (nitrile), a



halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0047] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0048] In order to improve the stability of a compound in the case of an uneasy carbonyl content compound with the inclination decomposed between iii arbitrary additive preservation or evaporation, an additional non-carbonyl ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of a carbonyl content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0049] Into the especially suitable precursive raw material mixture which consists of at least one carbonyl content compound Ru<sub>3</sub>CO<sub>12</sub>; Fe (CO) 5; Co<sub>2</sub> (CO) 8; Ru (CO) 3 ; Os<sub>3</sub>CO<sub>12</sub>; Cr (1, 3-cyclohexadiene) (CO) 6; CpCo (CO) 2; Mn<sub>2</sub> (CO) 10; CpMn (CO) 3; Mo (Cycloheptatriene) (CO) 3; Mo (CO) 6; nickel (CO) 4; Re<sub>2</sub> (CO) 10; CpRe (CO) 3; CpRh (CO) -- 2; Ru<sub>3</sub>(CO)<sub>12</sub>; W(CO)<sub>6</sub>; CpV(CO) 4; CF<sub>3</sub> -- Co (CO) -- four -- ; P 2(cyclo OKUTA screw-on); [ t(CO)<sub>2</sub>(cyclo OKUTA screw-on); Ir(CO) ] (CO)<sub>4</sub>Fe[P(OCH<sub>3</sub>) 3]; (CO)<sub>4</sub>Fe [N(CH<sub>3</sub>) 3] And CoNO (CO)<sub>3</sub> is contained. Cp is cyclopentadienyl or alternative cyclo pen TAJINIERU, and the substitution of H substituent by the methyl which dissolves, emulsifies or floats in C5-C12 alkane liquid, ethyl, an isopropyl, n butyl, the 2nd butyl, the 3rd butyl, trimethylsilyl, or other analogous substituents is taken into consideration here.

[0050] Precursive raw-material mixture for an alkoxy content compound: The suitable precursive raw material mixture of an alkoxy content compound is constituted as follows.

i) -- MR<sub>1</sub>xR<sub>2</sub>yAz -- here -- M -- Li, Na, K, Rb, and Cs -- Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, It is the element chosen from the group containing aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are B, aluminum, Ga, In, As, Sb, Si, germanium, Ti, Zr, or Hf suitably. Alkoxy \*\*\*\* of R1 is siloxy. R2 Moreover, a hydride, an alkyl, ARUKENIRU, cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, An amide, imide, a hydrazide, a phosphide, a nitrosyl, nitril (nitril), It may be the ligand chosen from the group containing a nitrate, nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0051] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0052] It decomposes between iii arbitrary additive preservation or evaporation, or in order to improve the stability of a compound in the case of an uneasy alkoxy content compound with the inclination which carries out a rearrangement chemically, an additional alkoxide ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of an alkoxy content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0053] Into the especially suitable precursive raw material mixture which consists of at least one alkoxy content compound Butoxy one, OCH (CF<sub>3</sub>)<sub>2</sub>, OCM<sub>2</sub> (CF<sub>3</sub>), OCM<sub>2</sub> (CF<sub>3</sub>)<sub>2</sub>, OSi (CH<sub>3</sub>)<sub>3</sub>, OC (CH<sub>3</sub>)<sub>3</sub> and OC (SiMe<sub>3</sub>)<sub>3</sub>, or OC(CF<sub>3</sub>)<sub>3</sub>Li, Na, K, Rb, Cs, Fr, Cu, Ag, Au, Hg, or Tl; Tetramethoxy, Tetrapod ethoxy \*\* tetrapod isopropoxy, 3rd butoxy one of a tetrapod, tetrapod iso butoxy, Tetrapod butoxy, a tetrapod OCH(CF<sub>3</sub>) 2, a tetrapod OCM<sub>2</sub> (CF<sub>3</sub>), A tetrapod OCM<sub>2</sub>(CF<sub>3</sub>) 2, a tetrapod (CH<sub>3</sub>) OC 3, a tetrapod (SiMe<sub>3</sub>) OC 3, a tetrapod (CF<sub>3</sub>) OC 3, or tetrapod OSi(CH<sub>3</sub>)<sub>3</sub>Si, germanium, Sn, Pb, Ti, Zr or Hf; VO (isopropoxy)<sub>3</sub>, triisopropoxy, 2nd butoxy one of TORI, TORI n butoxy, TORIISO butoxy, trimethoxy, TORIETOKISHI, TORI OCH(CF<sub>3</sub>) 2, TORI OCM<sub>2</sub> (CF<sub>3</sub>), TORI OCM<sub>2</sub>(CF<sub>3</sub>) 2, TORI (CH<sub>3</sub>) OC 3, TORI (SiMe<sub>3</sub>) OC 3, TORI (CF<sub>3</sub>) OC 3, or TORI OSi (CH<sub>3</sub>)<sub>3</sub> and B, aluminum, Ga, In, P, As, Sb; Et<sub>3</sub>Pb (isopropoxide), or (3rd butoxy) CuPMe<sub>3</sub> are contained, and these dissolve, emulsify or float in C5-C12 alkane liquid.

[0054] Precursive raw-material mixture for an amino content compound: The suitable precursive raw material mixture of an amino content compound is constituted as follows.

It Fr(s). i) --  $MR_1xR_2yAz$  -- here -- M -- Li, Na, K, Rb, and Cs -- Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, aluminum, It is the element chosen from the group containing Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are B, aluminum, Ga, In, As, Sb, Si, germanium, Sn, Pd, Zn, Cd, Hg, Ti, Zr, or Hf suitably. Those with amino \*\* and  $R_2 R_1$  Moreover, a hydride, an alkyl, ARUKENIRU, Cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, It may be the ligand chosen from the group containing nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if  $R_1$  and  $R_2$  are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0055] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0056] It decomposes between iii arbitrary additive preservation or evaporation, or in order to improve the stability of a compound in the case of an uneasy amino content compound with the inclination which carries out a rearrangement chemically, an additional amino ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of an amino content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0057] Into the especially suitable precursive raw material mixture which consists of at least one amino content compound Tetrakis (dimethylamino), Tetrakis Ti (dimethylamino) Zr, Hf, Si, germanium, Sn or Pb; diethylamino diethyl arsine; -- diethylamino arsine dichloride; -- bisdimethyl amino arsine chloride; --  $Me_2Zn(triethylamine)_2$ ; diethylamino dimethyl stannane; -- tris (dimethylamino) phosphine; -- tris Antimony; Tris (Dimethylamino) It Li(s). (Dimethylamino) arsine; -- tris (dimethylamino) stibine; -- tris screw (trimethylsilyl) erbium amide; -- screw (dimethylamino) (trimethyl ethyl ethylene diamino) aluminum; --  $(CO)_4Fe[N(CH_3)_3]$ ; -- Na or  $KN(SiMe_3)$ ; PENTA dimethylamino tantalum; -- diethylamino dimethyl tin; -- hexa dimethylamino JITANGU stainless steel -- Or tris dimethylamino (trimethylethylene diamino) titanium is contained, and these dissolve, emulsify or float in C5-C12 alkane liquid with an additional amine.

[0058] Precursive raw-material mixture for a phosphide content compound: The suitable precursive raw material mixture of a phosphide content compound is constituted as follows.

It Rb(s). i) --  $MR_1x(PR_2)_3yAz$  -- here -- M -- Li, Na, and K -- Cs, Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, It is the element chosen from the group containing Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are Cu, nickel, Pt, Ir, Cr, or Mo suitably. Moreover,  $R_1$  and  $R_2$  may be ligands chosen from the group containing a hydride, an alkyl, ARUKENIRU, cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, a hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if  $R_1$  and  $R_2$  are the same ligands, they may not be A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0059] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0060] It decomposes between iii arbitrary additive preservation or evaporation, or in order to improve the stability of a compound in the case of an uneasy phosphide content compound with the inclination which carries out a rearrangement chemically, an additional phosphide ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of a phosphide content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0061] Suitable precursive raw material mixture consists of  $CpCu(PEt_3)$ ,  $CpCu(triphenyl phosphine)$ ,  $CuPMe(3rd butoxy)_3$ ,  $Pt(PF_3)_4$ , nickel  $(PF_3)_4$  and  $Cr(PF_3)_6$ ,  $3(Et_3P)Mo(CO)_3$ , or  $Ir(PF_3)_4$ . Cp consists of cyclopentadienyl or alternative cyclo pen TAJNIERU, and the substitution of H substituent by the methyl which dissolves, emulsifies or floats in the C5-C12 alkane liquid containing a superfluous phosphine, ethyl, an isopropyl, n butyl, the 2nd butyl, the 3rd butyl, trimethylsilyl, or other analogous substituents is taken into consideration here.

[0062] Precursive raw-material mixture for a nitrate content compound: The suitable precursive raw material mixture of a nitrate

content compound is constituted as follows.

i) -- MR1xR2yAz -- here -- M -- Li, Na, K, Rb, and Cs -- Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, It is the element chosen from the group containing aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are Ti, Zr, Hf, Si, Ga, Sn, Co, V, or Cr suitably. R1 is a nitrate. R2 Moreover, a hydride, an alkyl, ARUKENIRU, Cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, It may be the ligand chosen from the group containing nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0063] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0064] It decomposes between iii arbitrary additive preservation or evaporation, or in order to improve the stability of a compound in the case of an uneasy nitrate content compound with the inclination which carries out a rearrangement chemically, an additional ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of a nitrate content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0065] Suitable precursive raw material mixture Ti (NO3)4, Zr (NO3)4, Hf (NO3)4, Si (CH3)3 (NO3), It consists of RuNO (NO3)3, a nitric-acid gallium, Sn (NO3)4, Co (NO3)3 and VO (NO3)3, or CrO2(NO3) 2, and these dissolve, emulsify or float in C5-C12 alkane liquid.

[0066] Precursive raw-material mixture for a halogenide content compound: The suitable precursive raw material mixture of a halogenide content compound is constituted as follows.

i) -- MR1xR2yAz -- here -- M -- Li, Na, K, Rb, and Cs -- Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, It is the element chosen from the group containing aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are Ti, Zr, Hf, Si, Ga, Sn, Co, V, or Cr suitably. R1 is a halogenide. R2 Moreover, a hydride, an alkyl, ARUKENIRU, Cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, It may be the ligand chosen from the group containing nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y \geq 0$ , and  $z \geq 0$ , and is the valence of the  $x+y=$  element M.

[0067] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0068] It decomposes between iii arbitrary additive preservation or evaporation, or in order to improve the stability of a compound in the case of an uneasy halogenide content compound with the inclination which carries out a rearrangement chemically, an additional halogenide ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of a halogenide content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0069] Suitable precursive raw material mixture is TiCl4, ZnCl2, ZrCl4, HfCl4, AlCl3, SiCl4, GaCl3, SnCl4, CoCl3, dimethyl -, diethyl -, or diisobutyl. - It consists of aluminum, B, germanium, Si, or As halogenide, and these dissolve, emulsify or float in C5-C12 alkane liquid.

[0070] Precursive raw-material mixture for a silyl content compound: The suitable precursive raw material mixture of a silyl content compound is constituted as follows.

i) -- MR1xR2yAz -- here -- M -- Li, Na, K, Rb, and Cs -- Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, It is the element chosen from the group containing aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi, and they are Ti, Zr, Hf, Si, Ga, Sn, Co, V, or Cr suitably. R1 is silyl. R2 Moreover, a hydride, an alkyl, ARUKENIRU, Cyclo ARUKENIRU, an aryl, an alkyne, a carbonyl, an amide, imide, A hydrazide, a phosphide, a nitrosyl, nitril (nitril), a nitrate, It may be the ligand chosen from the group containing nitril (nitrile), a halogenide, an azide, alkoxy \*\* siloxy, silyl and these halogenation, sulfonation, or a silicification derivative, and even if R1 and R2 are the same ligands, they

may not be so. A is arbitrary ligands which are chosen from a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, an alkyne, a hydrazine, a pyridine, a nitrogen heterocyclic compound, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, alkylidene, a nitrite, and the group containing water and by which coordinate bond is carried out. Moreover, it is  $x \geq 1$ ,  $y = 0$ , and  $z = 0$ , and is the valence of the  $x+y=$  element M.

[0071] ii) Inactive liquid inactive liquid is chosen from the group containing aliphatic hydrocarbon, an aromatic hydrocarbon, alcohol, the ether, an aldehyde, a ketone, an acid, a phenol, ester, an amine, alkyl nitril, a halogenated hydrocarbon, a silicification hydrocarbon, a thioether, an amine, a cyanate, an isocyanate, a thiocyanate, silicon oil, a nitro alkyl, alkyl nitrates, and such mixture. Suitably, inactive liquid consists of C5-C12 alkanes fundamentally.

[0072] It decomposes between iii arbitrary additive preservation or evaporation, or in order to improve the stability of a compound in the case of an uneasy silyl content compound with the inclination which carries out a rearrangement chemically, an additional silyl ligand may be added to mixture. In order to improve the stability of a compound in the case of the uneasy adduct of a silyl content compound, an additional adduct may be added to mixture. In order to improve the stability of a compound, other coordination compounds may be added to mixture and a phosphine, phosphite, an aryl, an amine, an arsine, a stibine, the ether, a sulfide, nitril, an isonitrile, an alkene, a pyridine, a heterocyclic compound, a tetrahydrofuran, a dimethylformamide, a large cyclic molecule, a Schiff base, a cycloalkene, alcohol, a phosphinic acid ghost, an alkyne, etc. are contained in them. All arbitrary additives occupy about 30% of inactive liquid by capacity.

[0073] suitable precursive raw material mixture --  $N(SiMe_3)_2$  -- Li, Na or K;  $B(CH_2SiMe_3)_3$ ;  $\{(Me_3Si)_2N\}_3B$ , aluminum and Ga, or In;  $(Me_3SiCH_2)_4-$  it consists of Ti, Zr or Hf;  $\{(Me_3Si)_2N\}_2Zn$ , and Cd or Hg, and these dissolve, emulsify or float in C5-C12 alkane liquid

[0074] The following examples are offered in order to illustrate this invention, and in order to prove some advantages of this invention.

[0075] Example 1: in the example of \*\*\*\*\* of the film within the chemical-vacuum-deposition reactor which uses precursive raw material mixture, a substrate is arranged in the suitable reactor for CVD, and it adheres to a compound element metal, metal oxide, a nitriding metal or the silicification metal Hf, and aluminum content film using two different precursive raw material mixture

[0076] Hafnium content precursive raw material mixture contains a 50g tetrakis (dimethylamino) hafnium and a 1g dimethylamine in a 1l. pentane. Aluminum content precursive raw material mixture contains a 50g trimethylamine and a 1g trimethylamine in a 1l. pentane. In this example, precursive raw material mixture is evaporated within a carburetor, and a steam is introduced in a CVD reactor. Precursive raw material mixture is 40 degrees C or 260 degrees C, and is suitably evaporated at 40 degrees C or 180 degrees C. Substrate temperature is about 100 degrees C or 1200 degrees C, and is 200 degrees C or 700 degrees C suitably. Since a metal membrane is adhered, reduction reaction agents, such as hydrogen, foaming gas, or such combination, are introduced. A suitable reducing agent is foaming gas. Since a metal oxide film is adhered, oxidizers, such as oxygen, ozone, water, a hydrogen peroxide, nitrous oxides, or such combination, are introduced. A suitable oxidizer is oxygen. Since a nitriding metal is adhered, nitriding reaction agents, such as ammonia, a hydrazine, a hydrogen azide, the 3rd butylamine, or such combination, are introduced. A suitable nitriding reaction agent is ammonia. Since a silicification metal membrane is adhered, silicification agents, such as a silane, a disilane, a chlorosilane, silylamine, and sill AZAN, are introduced in a CVD reactor together with a precursor steam, and a reducing agent is introduced in a CVD reactor together with a precursor steam. The steam and reaction agent of precursive raw material mixture are simultaneously introduced through a separate inlet port suitably.

[0077] This above-mentioned method is extended so that growth of the arbitrary compound element metal membranes and metal oxide film to which a chemical vacuum deposition adheres, a nitriding metal membrane, or a silicification metal membrane may be included using two or more different precursive raw material mixture or the precursive raw material mixture containing two or more precursors. Under the conditions that at least one element of a compound element film is drawn from precursive raw material mixture, this method is extended again so that growth of the arbitrary compound element metal membranes and metal oxide film to which a chemical vacuum deposition adheres, a nitriding metal membrane, or a silicification metal membrane may be included. You may adhere to other membranous elements using the precursive raw material which is not contained in the conventional bubbler technology or this invention. Using the precursive raw material mixture only containing one precursor, this above-mentioned method is extended again so that growth of the arbitrary single element metal membranes and metal oxide film to which a chemical vacuum deposition adheres, a nitriding metal membrane, or a silicification metal membrane may be included.

[0078] Example 2: in the example of film growth \*\*\*\*\* in which inactive liquid is not evaporated, inactive liquid is not evaporated but it is diverted from CVD or a ALD reactor Precursive raw material mixture consists of a precursor and inactive liquid, and inactive liquid is evaporated at an elevated temperature rather than a precursor. Precursive raw material mixture is introduced in a carburetor, and a precursor is evaporated there. Although inactive liquid is not evaporated, it is instead diverted from a reactor with the form of a liquid.

[0079] One of the possible equipment configurations is shown in drawing 30 . As shown in drawing 30 , the aforementioned raw material mixture is sent to a carburetor from ampul. Inactive liquid is not evaporated although the precursor in precursive raw material mixture is evaporated within a carburetor. While the evaporated precursor is transported into a reactor, the inactive liquid which is not evaporated flows out of a carburetor, and is collected in a trap. The temperature of a carburetor is set up lower than the boiling point of inactive liquid.

[0080] The precursive raw material mixture which consists of a dimethyl ethylamine alanine and Deccan (boiling point of 174 degrees C) is used for one suitable method under the carburettor temperature set as 90 degrees C.

[0081] This above-mentioned method is extended so that growth of the single element film to which it appears in a carburettor temperature volatilizing a precursor enough, and a chemical vacuum deposition or atomic-layer adhesion adheres under some

conditions of a low on the other hand rather than the boiling point of the inactive liquid in precursive raw material mixture, or a compound element film may be included.

[0082] Example 3: in the example of \*\*\*\*\* of the metal in an atomic-layer adhesion (ALD) reactor using precursive raw material mixture, metal oxide, or a nitriding metal, a substrate is arranged in the suitable reactor for ALD, for example, the F-200 reactor sold from a micro chemistry company, and it adheres to the compound element metal membrane containing Zr and Hf, a metal oxide film, or a nitriding metal membrane using two different precursive raw material mixture ALD is performed to circulating by sending the evaporated precursor, a reaction agent, and purge gas into a target in pulse by turns one by one.

[0083] Zirconium content precursive raw material mixture contains a 50g zirconium nitrate in a 1l. pentane. Hafnium content precursive raw material mixture contains the 50g 3rd butoxy hafnium of oxidization in a 1l. pentane. In this example, precursive raw material mixture is evaporated within a carburetor, and a steam is introduced in a reactor circulating. Since a metal membrane is adhered, reduction reaction agents, such as hydrogen, foaming gas, or such combination, are introduced. A suitable reducing agent is foaming gas. Since a metal oxide film is adhered, oxidizers, such as oxygen, ozone, water, a hydrogen peroxide, nitrous oxides, or such combination, are introduced. A suitable oxidizer is water. Since a nitriding metal is adhered, nitriding reaction agents, such as ammonia, a hydrazine, a hydrogen azide, the 3rd butylamine, or such combination, are introduced. A suitable nitriding reaction agent is ammonia.

[0084] Precursive raw material mixture is 40 degrees C or 260 degrees C, and is suitably evaporated at 40 degrees C or 180 degrees C. Substrate temperature is about 100 degrees C or about 1200 degrees C, and is 150 degrees C or 500 degrees C suitably. A precursor, a reaction agent, and inactive purge gas (N<sub>2</sub>, Ar, or other inert gas) are sent in pulse in a reactor in the following order.

1) the steamy 2 inactive purge gas of Hf content precursive raw material mixture -- the steamy 6 inactive purge gas 7 reaction agent 8 inactive purge gas [0085] of 3 reaction agent 4 inactive purge gas 5Zf content precursive raw material mixture a precursor and a reaction agent pulse (respectively the aforementioned steps 1 and 5 and Steps 3 and 7) -- 0.1 seconds -- or it continues for 0.5 seconds suitably for 1 second an inert gas purge pulse (Steps 2, 4, 6, and 8) -- 0.2 seconds -- or it continues for 2 seconds suitably for 5 seconds Step 1 or completion of 8 is equivalent to 1 cycle, and completion of 1 cycle adheres to about 0.4 or two monolayers of a ZrHf content film, i.e., about 0.1nm. In this example, since the suitable thickness of the ZrHf content film to which it adheres is 50nm, the above-mentioned gas exchange of 500 cycle is performed.

[0086] This above-mentioned method is extended so that growth of the arbitrary compound element metal membranes and metal oxide film to which atomic-layer adhesion adheres, a nitriding metal membrane, or a silicification metal membrane may be included using two or more different precursive raw material mixture or the precursive raw material mixture containing two or more precursors. Under the conditions that at least one element of a compound element film is drawn from precursive raw material mixture, this method is extended again so that growth of the arbitrary compound element metal membranes and metal oxide film to which atomic-layer adhesion adheres, a nitriding metal membrane, or a silicification metal membrane may be included. You may adhere to other membranous elements using the precursive raw material which is not contained in the conventional bubbler technology or this invention. Using the precursive raw material mixture only containing one precursor, this above-mentioned method is extended again so that growth of the arbitrary single element metal membranes and metal oxide film to which atomic-layer adhesion adheres, a nitriding metal membrane, or a silicification metal membrane may be included. In the alternative example, as mentioned above in Example 2, inactive liquid is not evaporated but it is diverted from a ALD reactor.

[0087] Example 4: in the example of \*\*\*\*\* of the silicification metal membrane within the atomic-layer adhesion reactor using precursive raw material mixture, a substrate is arranged in the suitable reactor for ALD, for example, the F-200 reactor sold from a micro chemistry company, and it adheres to a silicification cobalt film ALD is performed to circulating by sending the evaporated precursor, a reaction agent, and purge gas into a target in pulse by turns one by one. In this example, the hydrogen as a silicification agent is used for a silane as a reaction agent.

[0088] Cobalt content precursive raw material mixture contains Co<sub>2</sub>(CO)<sub>8</sub> [ 50g ] in a 1l. pentane. In this example, precursive raw material mixture is evaporated within a carburetor, and a steam is introduced in a reactor circulating.

[0089] Precursive raw material mixture is 40 degrees C or 260 degrees C, and is suitably evaporated at 40 degrees C or 180 degrees C. Substrate temperature is about 100 degrees C or about 1200 degrees C, and is 200 degrees C or 800 degrees C suitably. A precursor, a reaction agent, and inactive purge gas (N<sub>2</sub>, Ar, or other inert gas) are sent in pulse in a reactor in the following order.

1) Steamy 2 inactive purge gas 3 hydrogen 4 inactive purge gas 5 silane 6 inactive purge gas 7 hydrogen 8 inactive purge gas [0090] of Co content precursive raw material mixture a precursor and a reaction agent pulse (respectively the aforementioned steps 1 and 5 and Steps 3 and 7) -- 0.1 seconds -- or it continues for 0.5 seconds suitably for 1 second an inert gas purge pulse (Steps 2, 4, 6, and 8) -- 0.2 seconds -- or it continues for 2 seconds suitably for 5 seconds Step 1 or completion of 8 is equivalent to 1 cycle, and completion of 1 cycle adheres to about 0.4 or two monolayers of silicification cobalt, i.e., about 0.1nm. In this example, since the suitable thickness of the silicification cobalt film to which it adheres is 500nm, the above-mentioned gas exchange of 5000 cycle is performed.

[0091] This above-mentioned method is extended so that growth of arbitrary silicification metal membranes, such as CoSi<sub>2</sub>, HfSi<sub>2</sub> and NbSi<sub>2</sub> to which atomic-layer adhesion adheres, Pd<sub>2</sub>Si, PtSi, and TaSi<sub>2</sub>, TiSi<sub>2</sub>, VSi<sub>2</sub>, WSi<sub>2</sub>, ZrSi<sub>2</sub>, and arbitrary compound element silicification metals may be included, and atomic-layer adhesion uses at least one precursive raw material mixture, hydrogen or other reducing agents and a silane, or other silicification agents in order. A silane, a disilane, a chlorosilane, silylamine, silyl AZAN, etc. are contained in a silicification agent. In the another example, a silicification agent is introduced in precursive raw material mixture.

[0092] Example 5: in the example of copper \*\*\*\*\* , since this method adheres copper, it is used Copper content precursive raw material mixture contains 100g Cu (cyclopentadienyl) (PEt<sub>3</sub>) and 1g PEt<sub>3</sub> in a 1l. pentane. Precursive raw material mixture is

transported to a carburetor, it is evaporated at 60 degrees C there, and a steam is transported into a chemical-vacuum-deposition reactor. Reducing agents, such as hydrogen, exist in a reactor and it adheres to a copper film on the substrate suitably overheated by 100 degrees C or 300 degrees C there at 120 degrees C or 250 degrees C.

[0093] Example 6: in the example of \*\*\*\*\* of a tungsten, since this method adheres a tungsten, it is used Precursive raw material mixture contains a 100g hexacarbonyl tungsten in a 1l. hexane. Precursive raw material mixture is transported to a carburetor, it is evaporated at 80 degrees C there, and a steam is transported into a chemical-vacuum-deposition reactor. Reducing agents, such as hydrogen, exist in a reactor and it adheres to a tungsten film on the substrate suitably overheated by 200 degrees C or 700 degrees C there at 600 degrees C.

[0094] Example 7: in the example of \*\*\*\*\* of a tantalum nitride, since this method adheres a tantalum nitride, it is used Tantalum content precursive raw material mixture contains a 100g PENTA dimethylamino tantalum and 1g dimethylamino in a 1l. pentane. Precursive raw material mixture is transported to a carburetor, it is evaporated at 60 degrees C there, and a steam is transported into a chemical-vacuum-deposition reactor. Nitriding agents, such as ammonia, exist in a reactor and it adheres to a tantalum-nitride film on the substrate suitably overheated by 200 degrees C or 700 degrees C there at 500 degrees C.

[0095] Example 8: in the example of \*\*\*\*\* of an indium nitride, since this method adheres an indium nitride, it is used Indium content precursive raw material mixture contains 100g trimethylindium and a 1g dimethyl ethylamine in a 1l. pentane. Precursive raw material mixture is transported to a carburetor, it is evaporated at 60 degrees C there, and a steam is transported into a chemical-vacuum-deposition reactor. Nitriding agents, such as ammonia, exist in a reactor and it adheres to an indium-nitride film on the substrate suitably overheated by 100 degrees C or 700 degrees C there at 300 degrees C.

[0096] Example 9: in the example of \*\*\*\*\* of a zirconium silicate, since this method adheres a zirconium silicate, it is used Precursive raw material mixture contains the 100g 3rd butoxy zirconium of oxidization in a 1l. hexane. The 2nd precursive raw material mixture contains a 100g tetrapod ethoxy silane in a 1l. hexane. Precursive raw material mixture is transported to a carburetor, it is evaporated at 80 degrees C there, and a steam is transported into a chemical-vacuum-deposition reactor. Oxidizers, such as oxygen, exist in a reactor and it adheres to a zirconium-silicate film on the substrate suitably overheated by 200 degrees C or 700 degrees C there at 500 degrees C.

[0097] Example 10: \*\*\*\* of the aluminum2O3 gate dielectric of a transistor -- in this example, since this method adheres aluminum 2O3 which forms the gate dielectric layer 15 of PFET and NFET as shown in drawing 1 , it is used A silicon wafer substrate including patternizing structure is used. The selection field of a substrate is exposed including the \*\* trench separation (STI) oxide arranged between the sites of a transistor in the field where a transistor is arranged [ other fields ] for the selection field of Nakedness Si including a field oxide. Next, without arranging a substrate in a suitable modular cluster tool, and destroying a vacuum, the front face of Nakedness Si is processed, a SiOxNy layer is formed in a original position, and the thickness 2O3 layers of whose aluminum are 1nm or 100nm adheres according to the CVD process of this invention after that. Next, it adheres to a gate electrode on aluminum 2O3, without transporting a wafer to the 2nd module of a modular cluster tool, and destroying a vacuum there.

[0098] For adhesion of aluminum 2O3 by this method, precursive raw material mixture contains 103g dimethyl ethylamine Alan and a 10g dimethyl ethylamine in a 1l. hexane. Precursive raw material mixture is transported to a carburetor, it is evaporated at 80 degrees C there, and a steam is transported to the chemical-vacuum-deposition module of a cluster tool. Then, in a module, oxidizers, such as oxygen, ozone, N2O, water, or such mixture, exist, and it adheres to 2Oaluminum3 film at 200 degrees C.

[0099] Example 11: \*\*\*\* of the ZrO2 gate dielectric of a transistor -- in this example, since this method adheres ZrO2 which forms the gate dielectric layer 15 of PFET and NFET as shown in drawing 1 , it is used A silicon wafer substrate including patternizing structure is used. The selection field of a substrate is exposed including the \*\* trench separation (STI) oxide arranged between the sites of a transistor in the field where a transistor is arranged [ other fields ] for the selection field of Nakedness Si including a field oxide. Next, without arranging a substrate in a suitable modular cluster tool, and destroying a vacuum, the front face of Nakedness Si is processed, a SiOxNy layer is formed in a original position, and the thickness whose ZrO two-layer is 1nm or 100nm adheres according to the CVD process of this invention after that. Next, it adheres to a gate electrode on ZrO2, without transporting a wafer to the 2nd module of a modular cluster tool, and destroying a vacuum there.

[0100] For the adhesion of ZrO2 by this method, precursive raw material mixture contains the 100g 3rd butoxy zirconium of oxidization in a 1l. hexane. Precursive raw material mixture is transported to a carburetor, it is evaporated at 80 degrees C there, and a steam is transported to the chemical-vacuum-deposition module of a cluster tool. Then, in a module, oxidizers, such as oxygen, ozone, N2O, water, or such mixture, exist, and it adheres to ZrO2 film at 400 degrees C.

[0101] Example 12: \*\*\*\* of the gate dielectric of a transistor -- in this example, since this method adheres the gate dielectric of a transistor, it is used As shown in drawing 4 , a transistor element includes the gate electrode 52 which is formed on the conductive field 51 and formed on the gate dielectric 53, and one pair of n type sources / drain fields 54 which are formed in the both sides of the gate electrode 52. It adheres to the gate dielectric 53 by this method, and it consists of the dope of a different material or non-doping mixture, layers, or such combination. The arbitrary upper layers 57 of the gate dielectric 53 act as a dopant diffusion barrier, and stabilize structure between adhesion of the gate electrode 52. The arbitrary lower layers 55 of the gate dielectric 53 act as an electronic barrier layer or a layer which prevents oxidization of lower silicon, or play both roles. The interlayer 56 of the gate dielectric 53 is a dielectric layer which has high K.

[0102] The suitable lower layer 55 consists of dielectric materials, such as SiO2, SiOxNy, and Si3N4, and is prepared by oxidization or nitriding of a silicon substrate, or it adheres to it separately. A metal oxide metallurgy group silicate is contained in other suitable lower layer material. The middle dielectric layer 56 which has high K consists of the following dielectric materials 2O5, i.e., Ta, TiO2, ZrO2, HfO2 and aluminum 2O3, La2O3, Y2O3, an yttrium, an ulmin acid yttrium, an ulmin acid lanthanum, a silicic-acid lanthanum, a silicic-acid yttrium, a silicic-acid hafnium, silicic-acid zirconiums, these dopes or non-doping mixture, a layer, or



combination. An interlayer 56 is a layer which is a layer of the oxidation hafnium which could consist of some layers of a different material, for example, was inserted between the layers of an aluminum oxide, or consists of comparatively homogeneous mixture, such as mixture of a zirconium oxide and an oxidation hafnium. The arbitrary upper layers 57 are an interlayer's oxidation or nitriding front faces, or it adheres to dielectric materials, such as SiO<sub>2</sub>, SiO<sub>x</sub>N<sub>y</sub>, Si<sub>3</sub>N<sub>4</sub>, ZrO<sub>2</sub>, HfO<sub>2</sub> and aluminum 2O<sub>3</sub>, an aluminosilicate, a silicic-acid yttrium, a silicic-acid zirconium, a silicic-acid hafnium, silicic-acid lanthanums, these dopes or non-doping mixture, a layer, or combination. A suitable gate dielectric consists of a lower layer of SiO<sub>x</sub>N<sub>y</sub>, aluminum 2O<sub>3</sub> and the interlayer of ZrO<sub>2</sub> or HfO<sub>2</sub>, and an up barrier layer of nitriding metal oxide. It adheres to at least one component in the gate dielectric 53 by this method, i.e., the method of using precursive raw material mixture, evaporating precursive raw material mixture, and adhering a film with the steam of precursive raw material mixture in CVD or a ALD reactor.

[0103] Drawing 6 or drawing 13 is the cross section showing suitable formation of the transistor by this method. Formation of the gate dielectric 53 is performed in a original position within the cluster tool manufactured by applied MATERIARUZU. In drawing 6, the silicon substrate 50 which has a clean surface (natural SiO<sub>2</sub> does not exist) serves as a starting point. In drawing 7, a silicon substrate 50 is oxidized and nitrided and a SiO<sub>x</sub>N<sub>y</sub> layer is formed as a lower layer 55. In drawing 8, the interlayer 56 of a zirconium oxide is formed by this method. This adheres ZrO<sub>2</sub> film at 400 degrees C on a SiO<sub>x</sub>N<sub>y</sub> layer in the chemical-vacuum-deposition reactor in which the t-th butoxy zirconium of oxidation and the precursive raw material mixture of a hexane are used, and precursive raw material mixture is evaporated at 80 degrees C, next oxidizers, such as oxygen, ozone, N<sub>2</sub>O, H<sub>2</sub>O, or such mixture, exist. In drawing 9, the upper layer 57 is formed of plasma nitride formation of ZrO<sub>2</sub> front face. In drawing 10, it adheres to contest polysilicon as a gate electrode 52. In drawing 11 or drawing 13, the gate is formed, extended placing is performed, a spacer 58 is formed, the source / drain placing is performed and a perfect element is formed. The step of the continuing contact formation etc. is skipped.

[0104] Example 13: \*\*\*\* of the transistor using the metal gate -- in this example, in order that this method may form the transistor which uses the metal gate, it is used As shown in drawing 1, a CMOS integrated circuit contains both n type field-effect transistor (NFET) formed on a single substrate, and p type field-effect transistor (PFET). The NFET element 11 includes the gate electrode 14 which is formed on p type conductivity field 13 of a substrate 10, and is formed on the gate dielectric 15, and one pair of n type sources / drain fields 16 which are formed in the both sides of the longitudinal direction of the gate electrode 14. The gate electrode 19 by which similarly the PFET element 17 is formed on n type conductivity field 18 of a substrate 10, and is formed on the gate dielectric 15, and one pair of p type conductivity sources / drain fields 20 which are formed along with the side attachment wall of the both sides of the gate electrode 19 are included. NFET and a PFET element are separated by the \*\* trench separation 21 and the spacer 22. In this example, the gate electrodes 14 or 19 consist of the bulk metal or alloy which has a suitable work function. aluminum, Ag, Bi, Cd, Fe, Ga, Hf, In, Mn, Nb, Y, Zr, these alloys, etc. are contained in a metal suitable as a gate electrode 14 of the NFET element 11. At least one suitable metal for NFET is alloyed with W, Mo, Cr, and Cu, and may form the gate electrode 14. nickel, Pt, Be, Ir, Te, Re, Rh, etc. are contained in a metal suitable as a gate electrode 19 of the PFET element 17. At least one suitable metal for PFET is alloyed with W, Mo, Cr, and Cu, and may form the gate electrode 19. In this example, it adheres to at least one of NFET or the PFET element 14, i.e., a gate electrode, the gate electrode 19, and the gate dielectrics 15 by this method. As Example 2 described, it may adhere to the gate dielectric 15 by this method.

[0105] Example 14: \*\*\*\* of a transistor -- in this example, in order that this method may form the transistor which uses the metal gate, it is used Drawing 14 shows the general element structure containing NFET and PFET. In this example, NFET and PFET are formed on single p type electric conduction substrate 60. The NFET element 61 includes the gate electrode 62 which is formed on p type conductivity field of a substrate 60, and is formed on the gate dielectric 64, and one pair of n type sources / drain fields 65 which are formed in the both sides of the longitudinal direction of the gate electrode 62. Similarly, it is formed on n type well 72 of a substrate 60, the \*\* trench separation 73 dissociates, and the PFET element 71 includes the gate electrode 74 formed on the gate dielectric 64, and one pair of p type conductivity sources / drain fields 76 which are formed along with the side attachment wall of the both sides of the gate electrode 74.

[0106] Drawing 15 or drawing 24 is the fragmentary sectional view showing one example of the possible formation sequence of a standard CMOS flow. In drawing 15, the silicon substrate 60 which has the \*\* trench separation (STI) 73 and the n well 72 is formed in a part of p type electric conduction substrate. In drawing 16, the gate dielectric 64 and the masking layer (for example, Si<sub>3</sub>N<sub>4</sub>) 77 are formed on a substrate. As Example 12 described, it may adhere to the gate dielectric 64 by this method. Opening is formed in the portion which corresponds in the masking layer 77 at the NFET gate in drawing 17. The NFET gate electrode 62 is formed in drawing 18. Contest polysilicon, W, Mo, Ti, Cr, Cu, Fe, Mn, Nb, V, Re, Pt, Ag, Bi, Cd, Fe, Ga, Hf, In(s), Mn, Y and Zr, these dopes or a non-doping alloy, mixture, and a multilayer are contained in a material suitable as a NFET gate electrode 62. the NFET gate electrode 62 adheres by this method -- also having -- it is good In drawing 19, a chemical machinery polish (CMP) step is performed and flattening of the front face is carried out. In drawing 20 or drawing 22, the step shown by drawing 17 or drawing 19 is repeated for formation of a PFET element. Opening is formed in the portion which corresponds in the masking layer 77 at the PFET gate in drawing 20. The PFET gate electrode 74 is formed in drawing 21. Contest polysilicon, nickel, W, Mo, Ti, Cr, Te, Cu, Pd, Pt, Be, Au(s), Ir(s), Te and Rh(s), these dopes or a non-doping alloy, mixture, and a multilayer are contained in a material suitable as a gate electrode 74 of PFET. the PFET gate electrode 74 adheres by this method -- also having -- it is good In drawing 22, a chemical machinery polish (CMP) step is performed and flattening of the front face is carried out. In drawing 23, extended placing is performed through an obstruction to each of NFET and PFET. In drawing 24, the source / drain placing is performed through an obstruction after a spacer formation process to each of NFET and PFET, and element formation is completed. In this example, it adheres to the component of at least one or more layers, such as a gate electrode and a gate dielectric, or a layer by this method, i.e., the method of using precursive raw material mixture, evaporating precursive raw material mixture, and adhering a film



with the steam of precursive raw material mixture in CVD or a ALD reactor.

[0107] Example 15: \*\*\*\* of a multilayer capacitor -- in this example, the integrated circuit capacitor incorporating at least 1 component to which it adheres by this method is formed As shown in [drawing 2](#) , a typical capacitor is formed together with a barrier 32 on a substrate 30, and is connected to a transistor by the plug 31. A capacitor contains the bottom electrode 33, a dielectric (a ferroelectricity or non-ferroelectricity) 34, and the top electrode 35. In this example, it adheres to a plug 31, a barrier 32, the bottom electrode 33, a dielectric 34, and at least one capacitor element containing the top electrode 35 by this method.

[0108] [Drawing 25](#) or [drawing 29](#) is the fragmentary sectional view showing one example of the possible formation sequence of a capacitor. The substrate 30 which has a trench is formed in [drawing 25](#) . Substrates 30 are a silicon content semiconductor substrate, a silicon-on-insulator substrate, germanium substrate, a SiGe substrate or a GaAs substrate and other analogous substrates, a dielectric, a metal, an organic substrate, glass, a metallic oxide, plastics polymer substrates and these mixtures, combination, a multilayer, etc. In [drawing 26](#) , the plug material 31 and the arbitrary barriers 32 are formed. Plug material consists of the conventional electrical conducting materials, such as contest polysilicon, and W, Mo, Ti, Cr, Cu, and it may adhere to it using this method. The arbitrary electric conduction barriers 32 TaN, TaSiN, TiAlN, TiSiN, TaWN, TiWN, TaAlN, NbN and ZrN, TaTiN, IrO<sub>2</sub>, SiC, TiPt, TiNpt, TiAlN-Pt, Ru and RuO<sub>2</sub>, RuPt, Contest polysilicon of RuO<sub>2</sub>, WSi, Ti, TiSi(s), these dopes, or not doping, Or it consists of the conventional electrical conducting materials, such as aluminum, Pd, Ir, IrOx, Os, OsOx, MoSi, TiSi, ReO<sub>2</sub>, such mixture, or a multilayer, and may adhere using this method. The bottom electrode 33 is formed in [drawing 27](#) . The bottom electrode 33 consists of electrical conducting materials, such as nickel, Pd, Pt, Cu, Ag, Au, Ru, Ir, Rh, IrOx, RuOx, TaN, TaSiN, Ta, SrRuO<sub>3</sub> and LaSrCoO<sub>3</sub>, these dopes or a non-doping alloy, mixture, or a multilayer. A dielectric 34 is formed in [drawing 28](#) . A dielectric 34 consists of arbitrary insulating materials, such as metallic oxides, such as SiO<sub>2</sub>, SiOxNy, Si<sub>3</sub>N<sub>4</sub>, and Ta<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub>, HfO<sub>2</sub> and aluminum<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, these alloys, mixture, or a multilayer. Or a dielectric 33 may consist of compound element metallic oxides, such as a perovskite type oxide which has a chemical formula ABO<sub>3</sub>. B is at least one acidic oxide containing the metal chosen from the group containing aluminum, Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, and Cu here, and A is the cation of at least one addition which has the positive formal charge of about 1 or about 3. Silicates, such as strontium-titanate barium, zirconate, HAFUNETO (hafnate), a lead titanate, an ulmin acid yttrium, an ulmin acid lanthanum and titanic-acid zirconium lead, and a silicic-acid hafnium, a silicic-acid zirconium, the silicate with which rare earth was doped further are contained in such a compound element metallic oxide. The top electrode 35 is formed in [drawing 29](#) . The top electrode 35 consists of electrical conducting materials, such as contest polysilicon, nickel, Pd, Pt, Cu, Ag, Au, Ru, Ir and Rh, IrOx, RuOx, TaN, TaSiN, Ta, SrRuO<sub>3</sub> and LaSrCoO<sub>3</sub>, these dopes or a non-doping alloy, mixture, or these multilayers. Even if the top electrode and the bottom electrode are similar, they may not be so. In this example, it adheres to a plug, a barrier, a bottom electrode, a dielectric, and at least one or more layers containing a top electrode by this method, i.e., the method of using precursive raw material mixture, evaporating precursive raw material mixture, and adhering a film with the steam of precursive raw material mixture in CVD or a ALD reactor.

[0109] Example 16: \*\*\*\* of a trench capacitor -- in this example, the integrated-circuit trench capacitor incorporating at least one component to which it adheres by this method is formed One example which forms a trench capacitor on a substrate 30 is shown in [drawing 31](#) . The depression of a capacitor is formed in a substrate 30 and this is connected to a lower circuit through a plug 31. A circuit is covered by the dielectric insulating layer (separation dielectric) 83. Substrates are a silicon content semiconductor substrate, a silicon-on-insulator substrate, germanium substrate, a SiGe substrate, a GaAs substrate or other analogous substrates, a dielectric, a metal, an organic substrate, glass, a metallic oxide, plastics polymer substrates and these mixtures, combination, a multilayer, etc. Metallic oxides, such as SiO<sub>2</sub>, SiOxNy, Si<sub>3</sub>N<sub>4</sub>, silicic-acid Lynn glass, or aluminum<sub>2</sub>O<sub>3</sub>, were doped, or the dielectric insulating layer (separation dielectric) 83 is chosen from arbitrary insulating materials, such as non-doping mixture or these multilayers. On a plug and the depression of a capacitor, it adheres to the arbitrary electric conduction barrier layers 32, the pars-basilaris-occipitalis electrode layer 33, a dielectric layer 34, the top electrode layer 35, and the arbitrary dielectric buffer coats 36 one by one. Plug material consists of conventional material, such as contest polysilicon, and W, Mo, Ti, Cr, Cu, and it may adhere to it using this method. The arbitrary electric conduction barrier layers 32 TaN, TaSiN, TiAlN, TiSiN, TaWN, TiWN, TaAlN, NbN and ZrN, TaTiN, IrO<sub>2</sub>, SiC, TiPt, TiNpt, TiAlN-Pt, Ru and RuO<sub>2</sub>, RuPt, It consists of the conventional electrical conducting materials, such as RuO<sub>2</sub>, WSi, Ti, TiSi(s), these dopes, contest non-doping polysilicon or aluminum, Pd and Ir, IrOx, Os, OsOx, MoSi, TiSi, ReO<sub>2</sub>, such mixture, or a multilayer, and may adhere using this method. The pars-basilaris-occipitalis electrode 33 consists of electrical conducting materials, such as nickel, Pd, Pt, Cu, Ag, Au, Ru, Ir, Rh, IrOx, RuOx, TaN, TaSiN, Ta, SrRuO<sub>3</sub> and LaSrCoO<sub>3</sub>, these dopes or a non-doping alloy, mixture, or a multilayer. A dielectric 34 consists of arbitrary insulating materials, and metallic oxides, such as SiO<sub>2</sub>, SiOxNy, Si<sub>3</sub>N<sub>4</sub>, and Ta<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub>, HfO<sub>2</sub> and aluminum<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, these alloys, mixture, or a multilayer is contained in them. Or a dielectric 33 may consist of compound element metallic oxides which have a chemical formula ABO<sub>3</sub>, such as a perovskite type oxide. B is at least one acidic oxide containing the metal chosen from the group containing aluminum, Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, and Cu here, and A is the cation of at least one addition which has the positive formal charge of about 1 or about 3. Silicates, such as a barium strontium titanate, zirconate, HAFUNETO, a lead titanate, an ulmin acid yttrium, an ulmin acid lanthanum, titanic-acid zirconium lead, and a silicic-acid hafnium, a zirconium silicate, the silicate with which rare earth was doped further are contained in such a compound element metallic oxide. The top electrode 35 consists of electrical conducting materials, such as contest polysilicon, nickel, Pd, Pt, Cu, Ag, Au, Ru, Ir and Rh, IrOx, RuOx, TaN, TaSiN, Ta, SrRuO<sub>3</sub> and LaSrCoO<sub>3</sub>, these dopes or a non-doping alloy, mixture, or a multilayer. Even if the top electrode and the pars-basilaris-occipitalis electrode are similar, they may not be so. The arbitrary dielectric buffer coats 36 consist of arbitrary insulating materials, such as metallic oxides, such as SiO<sub>2</sub>, SiOxNy, Si<sub>3</sub>N<sub>4</sub>, TiON, AlN, SiN and TiN, and Ta<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub>, HfO<sub>2</sub> and aluminum<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, these alloys, mixture, a multilayer, or a compound element metallic oxide. Flattening of the layer to which it adheres one by one is carried out, and a trench capacitor is formed in the depression of a capacitor. Next, it

adheres to the insulating passive state layer 37 and the dielectric layer 38 between layers, and barrier structure is formed on the trench capacitor within a capacitor depression. The insulating passive state layer 37 consists of arbitrary insulating materials, such as metallic oxides, such as SiO<sub>2</sub>, SiO<sub>x</sub>N<sub>y</sub>, Si<sub>3</sub>N<sub>4</sub>, TiON, AlN, SiN and TiN, and Ta<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub>, HfO<sub>2</sub> and aluminum<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, these alloys, mixture, a multilayer, or a compound element metallic oxide. The dielectric 38 between layers is chosen from arbitrary insulating materials, such as SiO<sub>2</sub>, SiO<sub>x</sub>N<sub>y</sub>, Si<sub>3</sub>N<sub>4</sub>, silicic-acid Lynn glass, and a dope of metallic oxides, such as aluminum<sub>2</sub>O<sub>3</sub>, non-doping mixture or these multilayers. Bahia is formed in barrier structure. It adheres to the diffusion-barrier layer 81 and the metalization layer 82 on barrier structure and Bahia. The diffusion-barrier layer 81 is WN, TiN, or TaN. The metalization layer 82 is chosen from arbitrary electrical conducting materials, such as aluminum, W, Mo, Ti, Cr or Cu(s), these dopes or a non-doping alloy, mixture, or a multilayer. In this example, it adheres to at least one of the trench capacitor components containing the dielectric between electric conduction barrier material, a pars-basilaris-ossis-occipitalis electrode, a dielectric, a top electrode, an insulating passive state layer, and layers, a diffusion-barrier layer, a separation dielectric, and a metalization layer by this method.

[0110] Example 17: \*\*\*\* of wiring structure -- in this example, the integrated-circuit wiring structure incorporating at least 1 component to which it adheres by this method is formed As shown in drawing 3, typical wiring structure is formed by \*\*\*\*\*ing 42 in a trench 41 and Bahia into a dielectric layer 43. A dielectric layer 43 is chosen from arbitrary insulating materials, such as SiO<sub>2</sub>, SiO<sub>x</sub>N<sub>y</sub>, Si<sub>3</sub>N<sub>4</sub>, silicic-acid Lynn glass, and a dope of metallic oxides, such as aluminum<sub>2</sub>O<sub>3</sub>, non-doping mixture or these multilayers. A metalization layer is patternized by a pellet scene, a double pellet scene process, or lithography and etching. The metal thin film wiring 44 and a dielectric layer 45 exist in the dielectric-layer 43 bottom, the former is chosen as it from arbitrary electrical conducting materials, such as aluminum, W, Mo, Ti, Cr or Cu alloy, mixture, or these multilayers, and the latter is chosen as it from arbitrary insulating materials, such as SiO<sub>2</sub>, SiO<sub>x</sub>N<sub>y</sub>, Si<sub>3</sub>N<sub>4</sub>, silicic-acid Lynn glass, and a dope of metallic oxides, such as aluminum<sub>2</sub>O<sub>3</sub>, non-doping mixture or these multilayers. A trench and Bahia are filled up with drawing 4 with barrier material and a wiring metal. Barrier material is WN, TiN, or TaN, and a wiring metal is chosen from arbitrary electrical conducting materials, such as aluminum, W, Mo, Ti or Cu(s), these dopes or a non-doping alloy, mixture, or a multilayer, here. In this example, it adheres to at least one of a dielectric layer, metal thin film wiring, barrier material, and the wiring structure components containing a wiring metal by this method.

[0111] Although this invention has been especially described in relation to a suitable example, without swerving from the meaning and the range of this invention, if it is this contractor, he sets in detail and can understand the being a gestalt and that an above-mentioned change and other above-mentioned change are possible. Therefore, this invention is not an above-mentioned exact gestalt and the above-mentioned thing limited in detail.

[0112] As a conclusion, the following matters are indicated about the composition of this invention.

[0113] (1) the precursive raw material mixture containing at least one precursor compound which dissolves, emulsifies or floats in inactive liquid -- it is -- the above -- even if few -- one precursor compound -- a chemical formula [a formula 3] It has MR<sub>1</sub>xR<sub>2</sub>yAz. M here Li, Na, K, Rb, Cs, Fr, Be, Mg, Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Ru, Os, Co, Rh, Ir, nickel, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, It is the element chosen from the group containing aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, P, Sb, and Bi. R<sub>1</sub> and R<sub>2</sub> A hydride, an alkyl, the alkenyl, the cyclo alkenyl, An aryl, an alkyne, a carbonyl, an amide, imide, a hydrazide, a phosphide, A nitrosyl, nitril (nitril), a nitrate, nitril (nitrile), A halogenide, an azide, alkoxy \*\* siloxy, silyl, and these halogenation,

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